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AN EMANATION SYSTEM FOR DETERMINING SMALL QUANTITIES OF RADIUM-226

Richard L. Blanchard

Public Health Service
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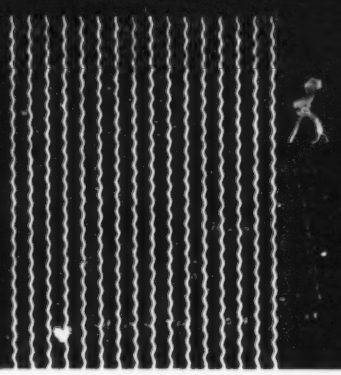

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U. S. DEPARTMENT OF HEALTH, EDUCATION, AND WELFARE

Public Health Service

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AN EMANATION SYSTEM FOR DETERMINING SMALL QUANTITIES OF RADIUM-226

by

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PREFACE

The necessity for evaluating health hazards associated with radium-226 has been recognized for a number of years. This requirement applies to environmental concentrations and to body burdens, whether in relation to the general population, or to specific occupational groups, or to communities located in areas containing certain geologic formations. The need for intensive studies on radium-226 has not declined, but has actually increased, as more knowledge has been accumulated and analyzed about this ubiquitous nuclide and its effects on health.

To date, most of the research concerned with the health hazards of radium-226 has been conducted at national laboratories, Federal laboratories, and universities. Clarification of its biological and environmental characteristics will undoubtedly lead to increased future surveillance by State and local health agencies.

This report explains in detail the construction and operation of equipment that has been found effective - with a high degree of specificity, sensitivity, and reliability - in the determination of radium-226. A small laboratory, or a laboratory with limited specialized facilities to deal with radioactivity and physical techniques, can attain a high level of proficiency in analysis for radium-226 by utilizing the procedural and explanatory material presented in this report.

ABSTRACT

A detailed account is given of the construction, operation, and characteristics of an emanation system for the determination of radium-226. The method is specific for radium-226, is well suited for large volume samples containing small quantities of this nuclide, and will accommodate any type of sample that can be prepared in an aqueous solution. The limit of detection at the 95-percent confidence level was calculated to be 0.016 picograms of radium-226, and the transfer and collection of radon-222 was determined to be 100 percent with an average deviation of 3 percent.

INTRODUCTION

The procedure described in detail in this report is based upon a method reported by Stehney (1) and Lucas (2). It is capable of measuring, with high accuracy, very small quantities of radium-226; it discriminates against other radium isotopes, as well as all other radioactive elements which may be present in the sample; it has a high degree of sensitivity and precision; and it produces accurate and dependable results.

Radium-226 is determined through its daughter, radon-222. The daughter is purged from a solution of the parent with an inactive gas and collected on activated charcoal at -78°C. The charcoal is then heated and the radon transferred to a zinc sulfide coated vessel where it is counted by scintillation techniques after radioactive equilibrium between radon and its daughters has been established.

The isotopes of radon which are transferred to the charcoal are: Radon-219, the daughter of radium-223, with a half life of 3.9 seconds; radon-220, the daughter of radium-224, with a half life of 54.5 seconds, and radon-222, the daughter of radium-226, with a half life of 3.82 days. Because of their short half lives, radon-219 and radon-220 do not reach the zinc sulfide counting vessel. Consequently, this method is specific for radium-226, and no corrections for the other isotopes of radium need be made.

The method described has a number of advantages over other methods, such as co-precipitation[1] or direct de-emanation into a counting vessel (4). The more important of these advantages are: Complete discrimination against all other radium isotopes without decay or energy measurements; complete discrimination against all other radioactive elements; and the possibility of assaying many types of samples with very little chemistry, which minimizes blank corrections of chemical reagents and eliminates the determination of chemical yields.

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[1] A paper by A. S. Goldin (3) provides an example of this methodology.

The system required to conduct this analysis is described below in detail. Each component of the system is evaluated and the overall efficiency, sensitivity, and reproducibility are determined.

THE VACUUM SYSTEM

The apparatus for the collection and transfer of radon is shown in figure 1. The system was constructed of stainless steel vacuum traps and 3/8-inch stainless steel type 304 seamless tubing with copper fittings. The de-emanation flask (D), the first water vapor trap (Σ_1), and the manometer (N) are constructed of Pyrex glass and connected to the system with Kovar seals. All metal connections were sealed with silver solder, with a number of joints included to enable the dismantling of the system by sections in case the need arose. Four vibration-eliminators were included to gain flexibility and to eliminate strain on the system while operating the vacuum pump.

This vacuum system construction proved to be quite durable and developed very few leaks with time and use. Following is a detailed description of the major components of the system. The letters after the various components refer to figure 1.

The Traps

The vacuum traps (B, E_2, E, and F) are constructed of 0.065-inch wall stainless steel tubing. Figure 2 is a diagram of the radon collection trap (F), containing 15 grams of activated coconut charcoal, 6-14 mesh, held in position with stainless steel gauze. The charcoal purification trap (B) is identical to (F) except that it has an overall length of 9 inches, a diameter of 1.9 inches, and contains 90 grams of activated coconut charcoal. The water vapor traps (E_2 and E) are also similar to (F) except they have the inlet at the top, the exit at the side, and are empty. The seams of the traps are arc-welded to eliminate the use of metals differing in their coefficients of expansion. The extreme temperature change (-78° to 450°C.) caused no leaks in these welded seams.

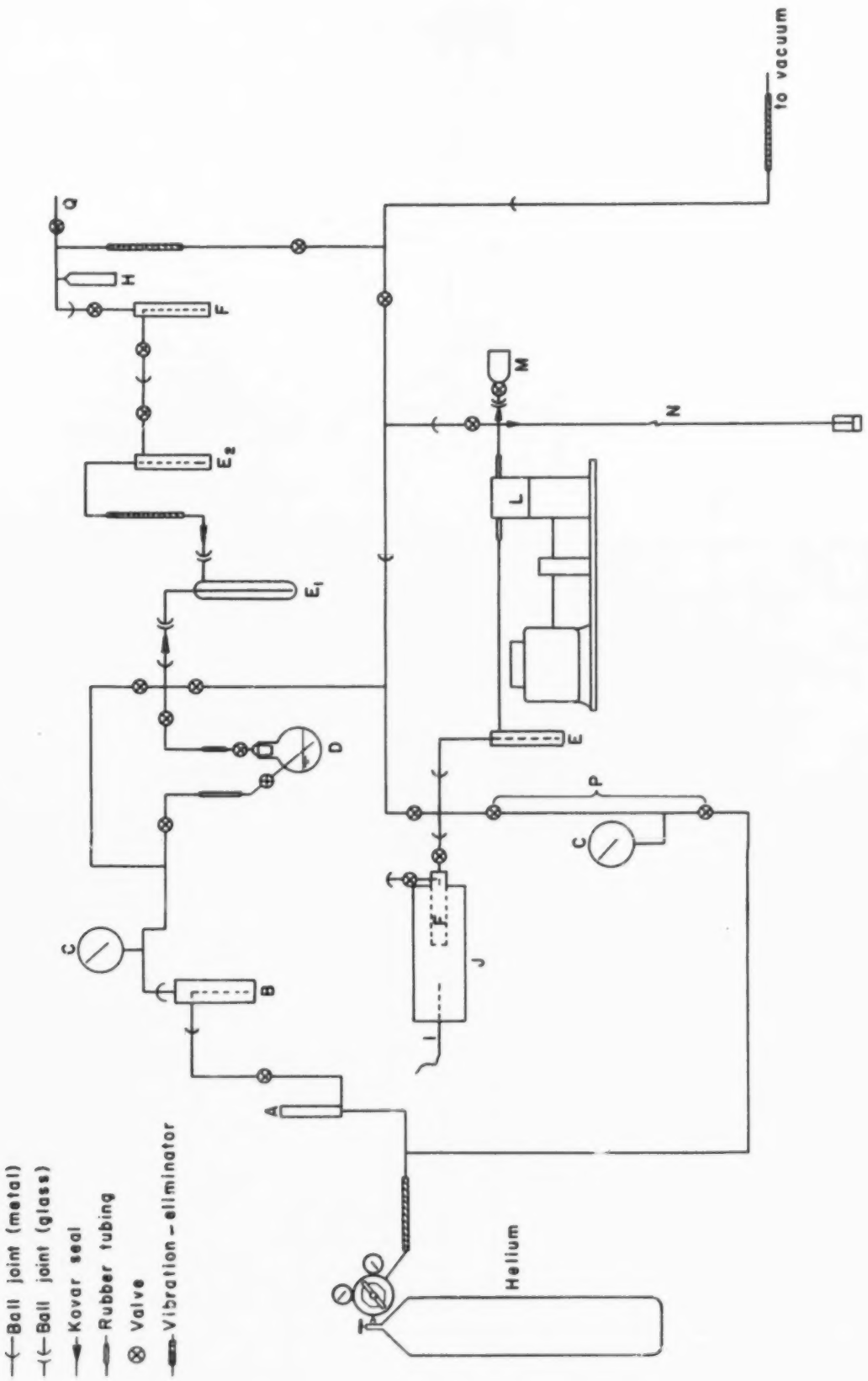

Figure 1. Radon Collection and Transfer System

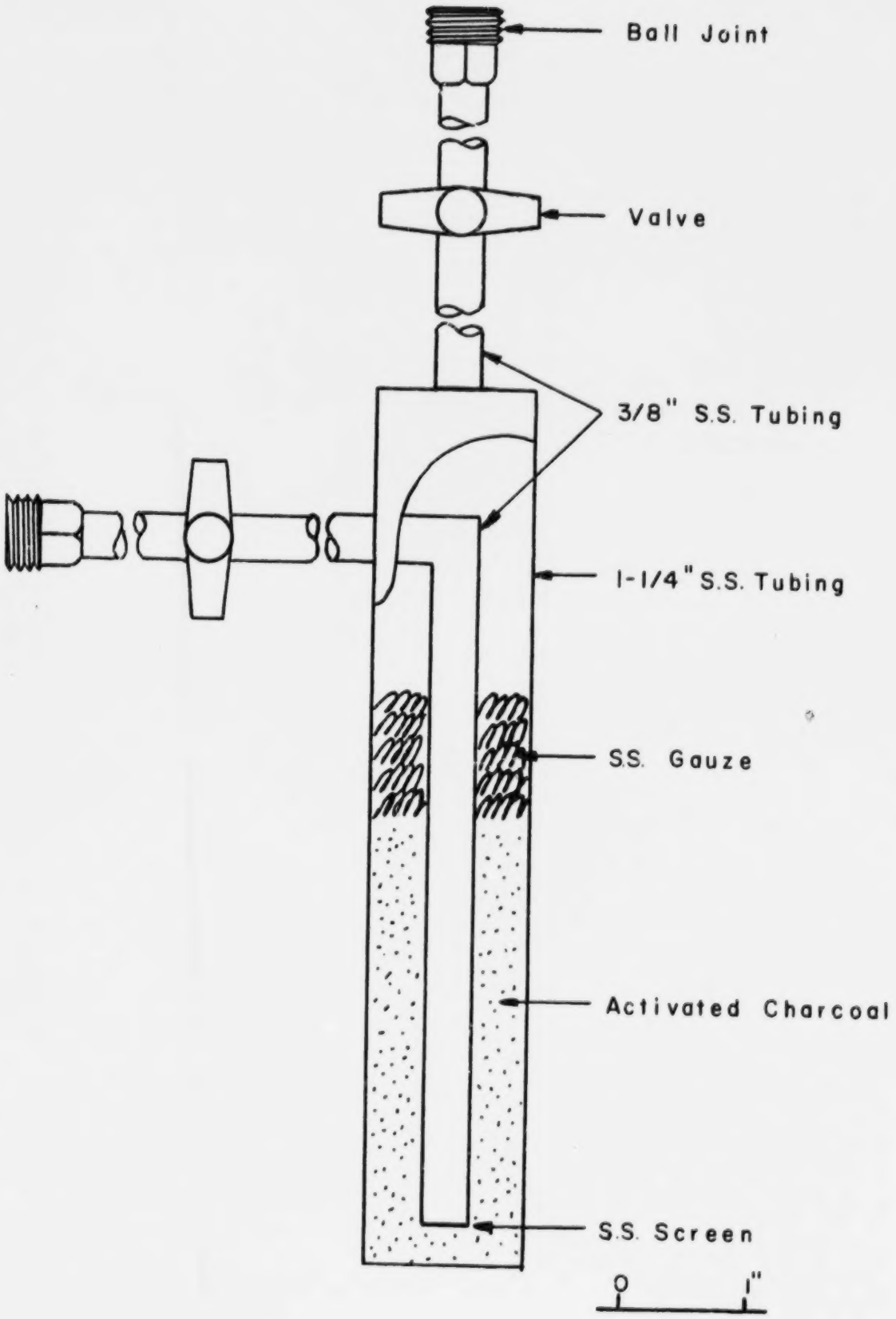

Figure 2. The Radon Collection Trap

The water vapor trap (E_1) is of Pyrex[2] glass, 35 by 250 millimeters, and contains 13 centimeters of 5-millimeter-diameter glass beads. The trap is held between two 9/18 Pyrex ball joints which are connected to the system with Kovar seals. Since this trap must be taken out at intervals because of the accumulation of water, a vibration-eliminator was included to permit easy removal.

The De-emanation Flask (D)

Figure 3 shows the de-emanation flask, a 500-milliliter Pyrex glass round-bottom flask with a 29/42 ground-glass neck. A 29/42 ground-glass adapter with a 4-millimeter vacuum stopcock is held firmly in the neck with two coiled spring clamps. A 10-millimeter outside-diameter Pyrex tube is sealed through the side of the flask and extends to within 5 millimeters of the bottom, where its tip is tapered to a 5-millimeter outside-diameter. Sealed to the other end of the tube, and outside of the flask, is a 4-millimeter vacuum stopcock. The flask is connected to the system by vacuum-pressure type rubber tubing which permits fast and easy connections. Thus, the flask as a unit can be de-emanated and removed from the system to await radon buildup and is easily accessible for cleaning through the ground-glass neck.

The Tube Furnace (J)

The tube furnace was constructed with a Chromel wire heating element (110 volts) wound around a 12- by 1½-inch cylindrical refractory. The unit is covered with half an inch of asbestos and three-quarters of an inch of pipe insulation. Asbestos cement was used to seal the ends of the furnace, leaving access through one end for the radon collection trap (F) and through the other end for the thermocouple (I). The furnace is regulated with an automatic indicator-controller.[3] Only 10 minutes are required to reach the operating temperature (450°C.).

The Manometer (N)

The manometer was constructed of Pyrex glass capillary tubing, 2-millimeter inside-diameter, sealed into a reservoir

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[2] Corning No. 96600

[3] Wheelco

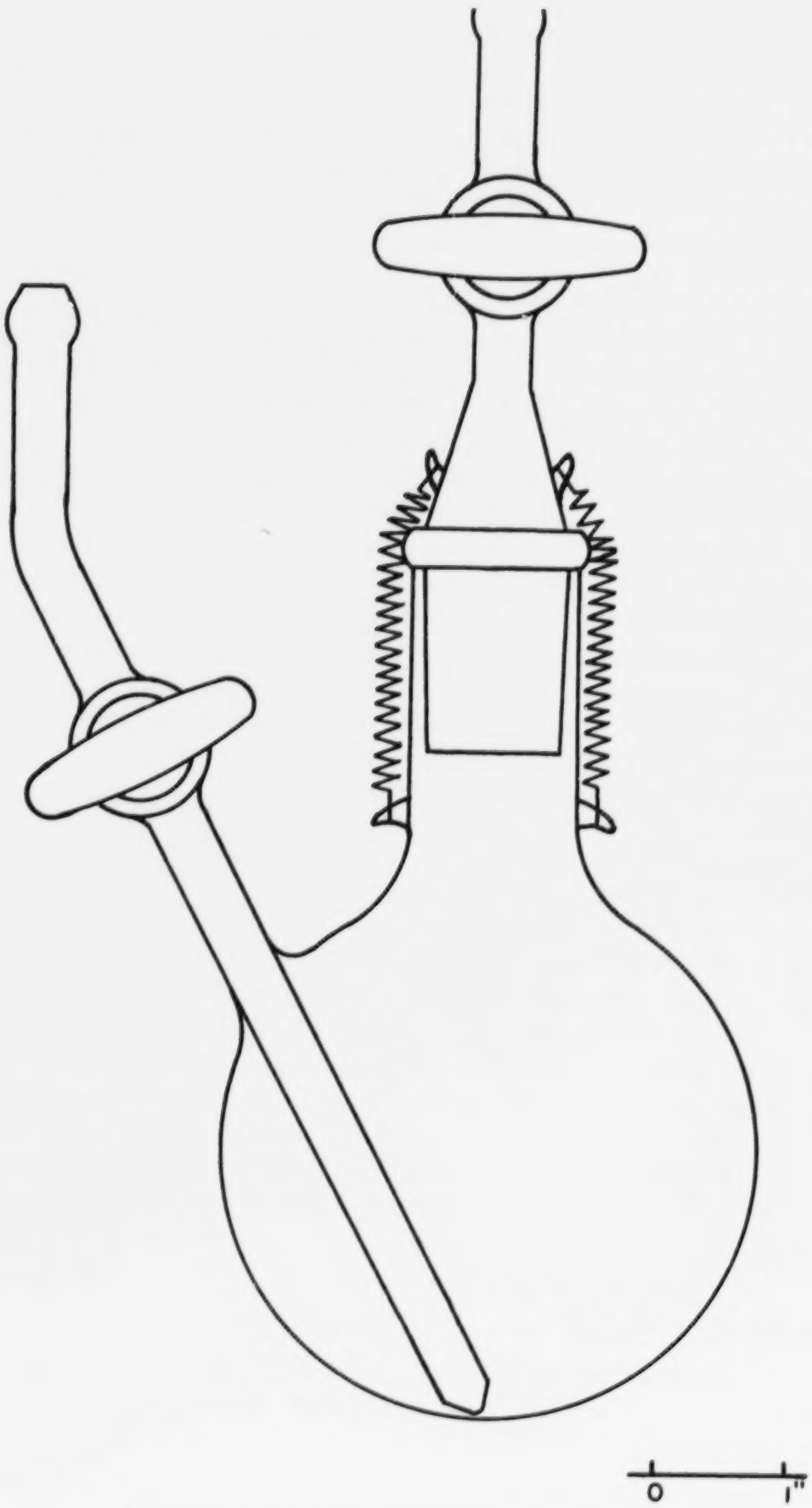

Figure 3. The De-emanation Flask

of double-distilled mercury. A small hole in the side of the reservoir above the mercury level equalizes the pressure with the atmosphere. The manometer is joined to the system with a Kovar seal.

The Flowmeter (A)

The flowmeter (No. 992 Hoke flowgage) has a maximum capacity of 1.7 liters per minute.

The Vacuum Gauge (H)

The vacuum gauge[4] is a thermocouple-type gauge, capable of measuring to 1 micron Hg pressure. It is operated with a thermocouple-gauge control.[5]

The Pressure-Vacuum Gauges (C)

These combination vacuum-pressure gauges measure both pressure (0-30 lb.) and vacuum (0-30 in.).

The Pump (L)

This is a Sigmamotor pump adapted for variable speed with a Zero-Max control. A pumping speed of approximately 0.1 liter per minute at one atmosphere (STP) was selected as the operating rate.

The Vacuum Pump

A vacuum pump,[6] with a 1/3-horsepower motor, supplies vacuum to the system.

The Radon Cell (M)

The radon scintillation cell used is the LAC-1.[7] This counter is described in detail by Lucas (2).

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[4] NRC Equipment Corp., Type 501

[5] NRC Equipment Corp., Type 701

[6] Welch Duo-Seal, No. 1405H

[7] Available from Wm. H. Johnston Laboratories, Inc.

THE RADON COUNTING SYSTEM

The system employed to count the alpha scintillations from the radon cell consists of a photomultiplier tube and preamplifier (which are housed in a light-tight enclosure), a stable high voltage supply, and a scaler.

The light-tight enclosure, 30 inches high, 15 inches wide, and 15 inches deep, was constructed of 1/4-inch plywood and lined on the inside with black felt. Access into the enclosure is provided through a 12- by 5-inch door which closes tightly against a rubber gasket, forming a light-tight seal.

The alpha scintillations from the radon cell are converted into electrical pulses by a photomultiplier tube,[8] powered by a model 300 beva laboratory 1.6-kilovolt power supply. The pulses from the phototube are fed through an emitter-follower to a Picker Labscaler. The emitter-follower was built by the electronics laboratory at Northeastern Radiological Health Laboratory.

The counting system draws its electrical power from a 110-volt line isolated from the main line supplying the rest of the laboratory. An A.C. voltage regulator[9] and an A.C. filter were installed in the line to gain a stable and low noise power-source.

The photomultiplier tube is operated at 830 volts on a plateau extending from approximately 700 volts to 950 volts. The background due to electronic noise is less than half a count per hour.

A plastic adapter into which the radon cell fits snugly was constructed to fit over the top of the photomultiplier tube. This assures a constant counting geometry.

PROCEDURE

Sample Preparation

As the purpose of this report is to describe a general method for the determination of radium-226 and not a specific method for any particular type sample, sample preparation

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[8] RCA 6655A

[9] Sorensen 5KVA

is not discussed in detail. It will be assumed that the sample, liquid or solid, has been adapted to the requirements of the procedure. Examples of samples which have been prepared for analysis without resorting to chemical separations are bone, marine carbonates, and sea water.

Preliminary De-emanation

The radium sample, in liquid form, is washed quantitatively into the 500-milliliter de-emanation flask and the volume adjusted to 100 to 300 milliliters. The ground-glass adapter with stopcock is secured in place and the flask is connected to the vacuum system. A coolant bath of dry ice and isopropyl alcohol is prepared in a Dewar flask and placed over the charcoal purification trap (B)(note 1). With the charcoal collection trap (F) out of the system, helium, 99.99 percent pure, is allowed to flow through the solution for 15 minutes at the rate of 1.5 liters per minute. At the completion of the de-emanation, the helium flow is stopped, the de-emanation flask is closed, and the time recorded (note 2). The sealed flask may now be removed from the system while awaiting radon buildup to allow continual use of the system (note 3).

The Collection of Radon

With the charcoal collection trap (F) in the system, the de-emanation flask containing the sample, previously de-emanated and sealed, is connected to the system. At this time, the collection system, except for the de-emanation flask, is evacuated and refilled with helium (note 4). Cooling baths (dry ice and isopropyl alcohol) are prepared in Dewar flasks and placed over the traps (B, E_1, E_2, and F) while a NaCl-ice bath is placed over the radium solution in the de-emanation flask (note 5). Helium is then allowed to flow at the rate of 1.5 liters per minute through the purification trap (B), the de-emanation flask (D), the water vapor traps (E_1 and E_2), the radon collection trap (F), and out through a valve (Q) to the atmosphere (note 6). After the de-emanation has proceeded for 15 minutes, the helium is shut off, the stopcocks on the de-emanation flask are closed, and the time is recorded (note 7).

At the completion of the de-emanation process, the inlet valve on the radon collection trap and the exhaust valve (Q) are closed while the valve leading to the vacuum pump is

opened. While at coolant-bath (dry ice and isopropyl alcohol) temperature, the radon collection trap is evacuated to 20 microns Hg (note 8). When the pressure reaches 20 microns Hg, the upper valve on the radon collection trap is closed. Atmospheric pressure is restored in the vacuum lines with helium, and the trap containing the radon is removed from the collection system and connected to the transfer system.

The Transfer of Radon

A Dewar flask containing a coolant bath (dry ice and isopropyl alcohol) is placed over the water vapor trap (E)(note 9). The tube furnace (J), preheated to 450°C., is placed over the radon collection trap and 5 minutes are allowed to insure a charcoal temperature of 450°C. During this heating time, the transfer system is flushed twice with helium and then evacuated.

The 15-milliliter helium reservoir (P) consisting of tubing between two valves is adjusted to one atmosphere with helium. The two valves leading to the main vacuum line are closed, and the valve on the radon collection trap is opened. The top valve on the helium reservoir is opened, and the 15 milliliters of helium are allowed to mix with the radon. After a 3-minute mixing period, the Sigmamotor pump (L) is started, and the helium-radon mixture is pumped to the radon cell (M). When the mercury in the manometer (N) becomes stationary, indicating that the pressure to the left of the Sigmamotor pump has reached the lower limit of the pump, another 15-milliliter increment of helium is supplied from the reservoir to the transfer system. This increment of helium, like the first, is completely removed to the radon cell side of the system before the next increment is introduced. This process is continued until the manometer indicates that the pressure on the right side of the pump is at one atmosphere. To reach one atmosphere requires six of the 15-milliliter transfers. Two minutes are allowed after the introduction of the second and third increments of helium to insure helium-radon mixing, during which time the Sigmamoter pump is off (note 10).

When the transfer is completed, the stopcock on the radon scintillation cell (M) is closed, and the tube-furnace is removed from the radon collection trap. Four hours are allowed to elapse between radon transfer and the counting of the sample to insure equilibrium between radon and its daughters.

During the transfer of the radon, another sample can be de-emanated and the radon collected on another charcoal trap. This allows a larger number of analyses to be performed in a day.

Notes

Note 1. Isopropyl alcohol is preferred to acetone as a cooling medium because the boiling action due to the escape of CO_2 is considerably less, since isopropyl alcohol becomes quite viscous at dry-ice temperature (its melting point is -85.8°C.).

Since radium may be present in the metal walls of the helium container, this trap eliminates any radon which may be present in the helium.

Note 2. This is the start of growth of radon-222 from its radon-free radium-226 parent.

Note 3. A helium atmosphere is maintained at all times within the system to prevent contamination from atmospheric radon and to protect the charcoal from adsorbable atmospheric components.

Note 4. The charcoal in the radon collection trap must be degassed before it is used for the first time and thereafter kept in a helium atmosphere. This is accomplished by heating to 450°C. under a vacuum for 1 hour.

Coconut charcoal contains approximately 0.013 picograms of radium-226 per gram (1). This radium-contaminated charcoal creates a background problem which can be quite severe when small quantities of radium are being determined. Therefore, shortly before the radium solution is de-emanated, the radon collection trap is degassed to eliminate any radon which has formed since the last emanation. This is accomplished by heating the charcoal trap to 450°C. for 20 minutes, accompanied by vacuum pumping. This step is especially important if the radon trap has not been used recently.

Note 5. The coolant bath reduces the evaporation of the solution during de-emanation and consequently decreases the corrosion of the valve and the line between the de-emanation flask and the first water vapor trap. This valve, incidentally, is a stainless steel valve with Teflon seals.

Note 6. For the de-emanation and collection of radon, helium is preferred to aged air as it is not adsorbed on the activated charcoal; whereas carbon dioxide, carbon monoxide, oxygen, nitrogen, and other gases are adsorbed to varying degrees. Valves leading to the transfer system must be closed during de-emanation.

Note 7. If another analysis is to be made of this sample, the growth time will begin at the end of the de-emanation period.

Note 8. Approximately 3 minutes are necessary to reach the required vacuum of 20 microns Hg. The loss of radon during this evacuation is less than 0.3 percent at the temperature of the coolant bath of dry ice and isopropyl alcohol.

Note 9. This trap will prevent any moisture which may be present from entering the radon scintillation cell.

Note 10. Since more than 99 percent of the radon is transferred to the radon-cell side of the pump after the third transfer, it is not necessary to allow time for radon-helium mixing before the fourth, fifth, and sixth transfers.

SYSTEM EVALUATION

Counter-System Backgrounds

The windows of the radon scintillation cells were constructed of Pyrex glass which give it a somewhat higher alpha background than counters with quartz windows. Nevertheless, the background with these cells has proved to be low and uniform. Initially the cell backgrounds were about 0.06 count per minute; after many months of continual service the backgrounds increased to about 0.15 count per minute.

The backgrounds of the radon collection and transfer systems were determined by operating the systems exactly the same as when a sample was being analyzed, except that the radium solution in the de-emanation flask was replaced with distilled water. The radon collected in the background determinations was derived from radium impurities in the coconut charcoal, the stainless steel tubing, and, possibly, from the

Pyrex glass in the system.[10] Very little difference could be detected when distilled water was used in the de-emanation flask or when the flask was bypassed. This would indicate an undetectable quantity of radium in the distilled water used to make up the radium solutions.

A very large difference in the background was noted when the radon trap was allowed to stand a few days between emanations and the collection system was not evacuated immediately before the de-emanation. For example, after the system had been idle for a few days and was not evacuated before the de-emanation, a radium equivalent content of 0.250 ± 0.020 picocurie was found. However, when the radon trap was degassed and the system was evacuated immediately before the de-emanation, a radium equivalent content of only 0.021 ± 0.003 picocurie was detected. This illustrates the necessity for degassing the radon collection trap and for evacuating and refilling the system with fresh helium prior to de-emanation, especially after the system has remained idle for any length of time.

The systems are never allowed to be open to the atmosphere. All valves leading to the atmosphere are kept closed except when opening is required by the procedure. When not in operation, the systems are maintained at one atmosphere with helium.

Counting Efficiency

The counting efficiency for each of the four available radon cells was determined using both purified air and helium as fillers. A solution of known radium-226 content, contained in a bubbler of the type described by Holaday, et al. (5), was de-emanated. The radon de-emanated from the bubbler was passed through a small tube containing asbestos impregnated with sodium hydroxide[11] and a drying agent $Mg(ClO_4)$ and into an evacuated radon cell. A de-emanation efficiency of 100 percent was verified by immediately repeating the de-emanation with a second radon cell. No alpha activity was detected in the second cell. After de-emanation and collection, the counters were allowed to stand 4 hours to insure equilibrium between the radon and radon daughters, and were then counted.

The results obtained by employing both helium and aged air as counter fills are given in table 1. These data illustrate

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[10] See note 4 in the previous section.

[11] Available as Ascarite from Arthur H. Thomas Company.

the reproducibility possible with these counters, and the almost identical efficiencies of the various counters. This high counting-reliability enhances the value of this method, especially when very small quantities of radium-226 are encountered.

The data listed in table 1 also illustrate the advantage of helium over air as a counting fill. The counting efficiencies are increased to 80 percent when helium is employed. The additional number of counts acquired by use of helium are quite important when dealing with small quantities of radium. The use of helium not only increases the sensitivity of the method but also improves the counting statistics which, in turn, decrease the uncertainty in the final counting rate.

Table 1. Efficiencies of the Radon Counters

Counter Number	Air				Helium			
	Trial #1	Trial #2	Trial #3	Average	Trial #1	Trial #2	Trial #3	Average
B-1101	69.8 ± 1.8	72.9 ± 1.5	70.4 ± 1.2	71.0 ± 1.5	78.8 ± 1.6	80.3 ± 1.8	81.0 ± 1.4	80.0 ± 1.6
B-1102	72.9 ± 1.5	71.7 ± 1.5	70.5 ± 1.6	71.7 ± 1.5	79.7 ± 1.8	79.1 ± 1.7	80.5 ± 1.7	79.8 ± 1.7
B-1103	70.3 ± 1.4	72.5 ± 1.5	71.2 ± 1.5	71.3 ± 1.5	80.3 ± 1.7	80.0 ± 1.5	78.0 ± 1.3	79.5 ± 1.5
B-1104	-	-	-	-	79.0 ± 1.7	82.5 ± 1.8	82.6 ± 1.5	81.4 ± 1.7

Note: Efficiencies are stated as percentages; uncertainties given are the standard deviation (σ).

The increased counting rate obtained with the use of helium should be expected since the density of air is 6.85 times that of helium, and the relative stopping power is about 0.35 for helium.[12] Consequently, the mean range of an alpha particle emitted from a radon atom (the lowest energy alpha emitted in the radon-radon daughter mixture) is increased from 4.05 centimeters in air to about 11.6 centimeters in helium. Therefore, in helium no alpha particles are out of range of the ZnS(Ag) phosphor in the counters used except those which impinge upon the Pyrex glass window.[13] This is not the case for a counter filled with air.

The Efficiency of the Transfer and Collection System

The efficiencies of the collection and transfer processes were determined by analyzing two radium-226 standards obtained from the National Bureau of Standards. These included

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[12] This is the relative stopping power for a 6.3 mev alpha particle in helium (6). Since the radon alpha has an energy of 5.486 mev, the relative stopping power for a radon alpha will differ slightly from the 0.35 value used.

[13] These bell-shaped cells have a diameter of 4.7 centimeters and a height of 6.4 centimeters.

a standard radium-226 solution from which a standard was prepared consisting of 1.00 picogram of radium, and a 20-gram standard limestone sample from which a standard containing 7.00 picograms of radium was prepared. The latter standard was dissolved in HNO_3 and both standards were placed in de-emanation flasks and diluted to 100 milliliters in 0.1N HNO_3.

The radium-226 concentrations determined for both standards and the overall efficiencies of the radon collection and transfer system are shown in table 2. The results of these analyses indicate that the collection of the radon on the charcoal and the transfer to the radon cell are very nearly 100 percent complete. The average recovery of all nine trials was 100 ± 4 percent with an average deviation of less than 3 percent, illustrating the accuracy and the reproducibility of the method.

Table 2. Standard Radium-226 Determinations

Trial No.	Standard	Ra^{226} in Standard (picograms)	Ra^{226} Recovered (picograms)	Percent Efficiency
1	Limestone	7.00	6.8 ± 0.3	97 ± 4
2	Limestone	7.00	6.8 ± 0.3	97 ± 4
3	Limestone	7.00	7.1 ± 0.3	101 ± 4
4	Limestone	7.00	6.7 ± 0.3	96 ± 4
5	Limestone	7.00	7.2 ± 0.3	103 ± 4
6	Solution	1.00	1.02 ± 0.04	102 ± 4
7	Solution	1.00	0.99 ± 0.03	99 ± 3
8	Solution	1.00	0.97 ± 0.03	97 ± 3
9	Solution	1.00	1.05 ± 0.04	105 ± 4

The efficiency with which radon is collected on the cooled charcoal was determined by varying the purging time and the volume of sample solution. Ninety-five percent of the radon was collected after purging for only 5 minutes at a rate of 1.5 liters per minute, while 100 percent was collected after

purging only 10 minutes at the same rate. The collection efficiency did not differ from sample volumes of 100 to 400 milliliters.

The efficiency for the transfer of radon from the charcoal to the radon counter was determined independently by making a second transfer, immediately following the first, into another radon cell. The alpha activity recovered in the second radon cell was less than 0.3 percent of that found in the first cell, indicating that the radon transfer process is highly efficient.

The limit of detection for the system is calculated to be 0.016 picogram of radium-226 at the 95 percent confidence level. The accuracy which can be attained is dependent upon the size of the sample available for analysis. For example, a liter water sample containing 0.001 picogram per milliliter can be determined within $\pm$ 4 percent, whereas a 20-gram solid sample containing 0.01 picogram per gram can be determined with less than a 10-percent uncertainty. Since these values were calculated on the basis of a 200-minute count, they can be reduced by increasing the counting time.

CALCULATIONS

The time allowed for the growth of radon into a freshly de-emanated radium solution depends upon the quantity of radium in the sample. There is no purpose in allowing the growth time to exceed a time sufficient to produce a statistically sound count rate of radon plus daughters. The rate at which radon accumulates from radium is given by Eq. (1)

$$A_{Rn} = A_{Ra}(1 - e^{-0.181t}) \qquad (1)$$

where A_{Rn} and A_{Ra} are the activities of radon-222 and radium-226, respectively, and t is the growth time in days. Thus, by knowing t, the time for radon growth, and measuring A_{Rn}, it is simple to calculate the quantity of radium-226 in a sample.

In practice, Eq.(1) is applied to determine the fraction of radioactive equilibrium between radium and radon (A_{Rn}/A_{Ra}) which exists at the time of de-emanation. In

addition, corrections must be applied to the count rate measured to obtain the correct disintegration rate of the radon present. These corrections include the counting efficiency (E) and background (B_1) of the counter, the background of the collection and transfer system (B_2), the radon decay that occurred between transfer and counting (D), and the excess volume not included in the radon cell on the right side of the Sigmamotor pump (V). By applying these corrections along with the fraction of the radium-radon equilibrium attained (G), as shown in Eq. (2), the radium-226 disintegration rate in the sample can be obtained.

$$A_{Ra226} = \frac{\text{counts/minute} - (B_1 + B_2)}{(E)\quad (D)\quad (V)\quad (G)\quad (3)} \qquad (2)$$

The activity measured is divided by 3 in Eq. (2) to correct for the two alpha-emitting daughter products of radon, polonium-218 and polonium-214, which are in equilibrium with radon at the time of counting, 4 hours after the transfer. It has been reported that the contribution of each alpha-emitting nuclide (radon and the two alpha-emitting daughters) to the total alpha activity is dependent upon the cell dimensions (7). For a radon cell of the dimensions used, employing helium as the fill, radon and each daughter will contribute equally to the total activity. Hence, radon is responsible for one-third of the alpha activity measured.

By dividing the disintegration rate per minute by 2.22, the result of Eq. (2) may be converted into picocuries, or, since the element is radium-226, picograms of radium-226.

SUMMARY

The construction and operation of a vacuum system for the analyses of radium-226 via radon-222 is described in detail. The system is especially suitable for large-volume samples containing small quantities of radium, and will accommodate any type of sample that can be prepared as an aqueous solution.

Two standard radium-226 samples were used to evaluate the method. The results of this evaluation showed this to be a highly reliable and accurate method for determination of small quantities of radium-226. Nine analyses were conducted on the two standard samples, with an average efficiency of 100 percent

and an average deviation of less than 3 percent. It was calculated that, with this system, 0.016 picogram of radium-226 can be detected at the 95-percent confidence level. In addition, it should be pointed out that this method is specific for radium-226, discriminating against all other isotopes of radium.

The efficiencies of the radon cells were determined using both purified air and helium as counter fills. It was found that the counting efficiencies were increased considerably by the use of helium instead of air.

The time required for the collection and transfer of the radon is approximately 1 hour. When analyzing continuously, however, the collection and transfer of the radon can be conducted simultaneously, thereby reducing considerably the time required per analysis.

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